Opportunities for Ultrafast x-ray physics at the APS

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Outline

- Some motivation for Ultrafast x-rays physics
- Brief survey of current and future sources (laser based vs SR based)
- Some Ultrafast X-ray Physics activities at the APS
- Ultrafast control of x-rays: transient optics
- Related Experiments
  - coherent phonon spectroscopy (inelastic scattering)
  - anomalous transmission and the Pendellösung effect
  - ambipolar diffusion and e-phonon coupling
- Scaling it towards femtoseconds

Ultrafast lasers produce large strain fields which cannot directly be probed by laser methods. The use of X-rays allow for a direct determination of strain and allow for element specific spectroscopies (EXAFS).
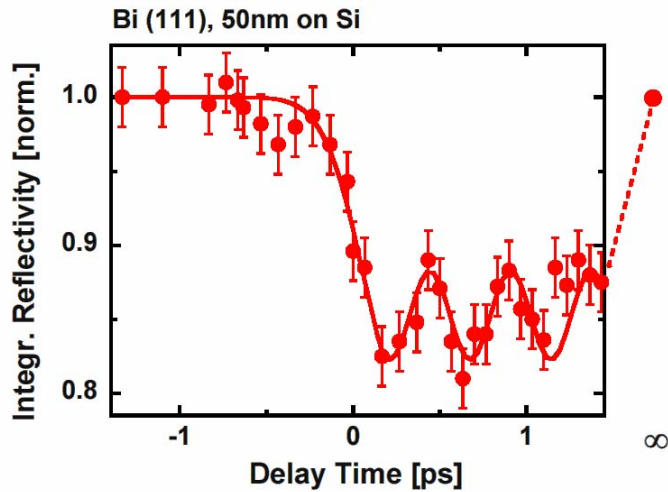
Some frontier questions:
•Do optical phonons play a role in structural phase transitions?
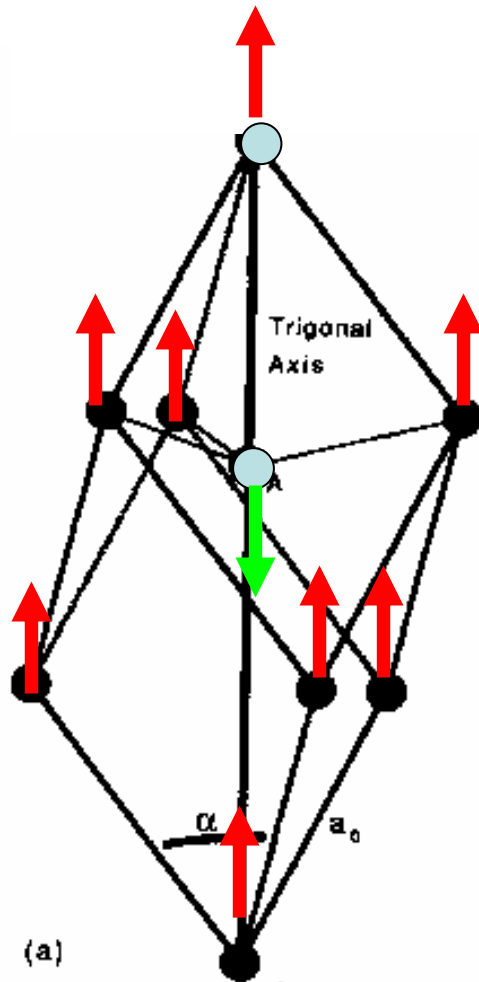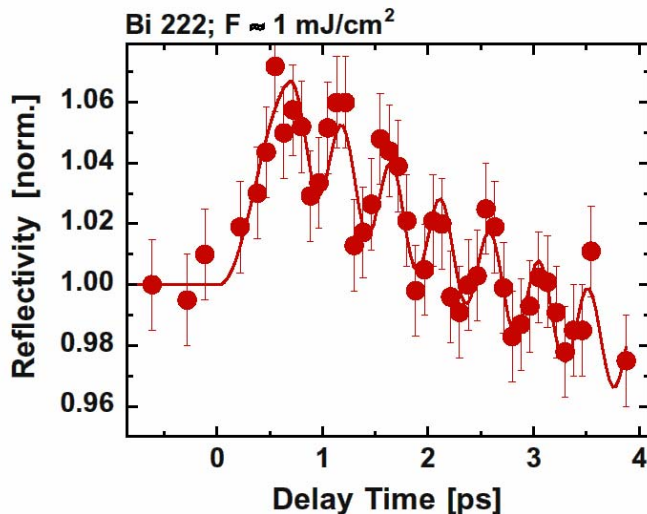•What is the nature of the observed mode softening?
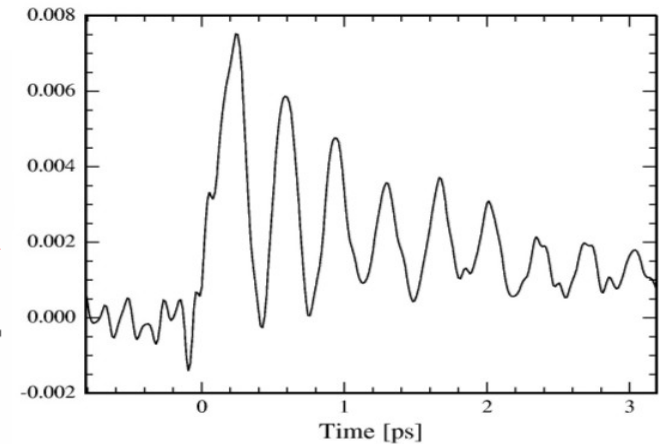
An example: coherent electron-phonon excitation.

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X-ray Diffraction

Bi (111), 50nm on Si

Sokolowski-Tinten *et al.,*
Nature **422** , 287 - 289 (2003)

Bi 222; F ≈ 1 mJ/cm^2
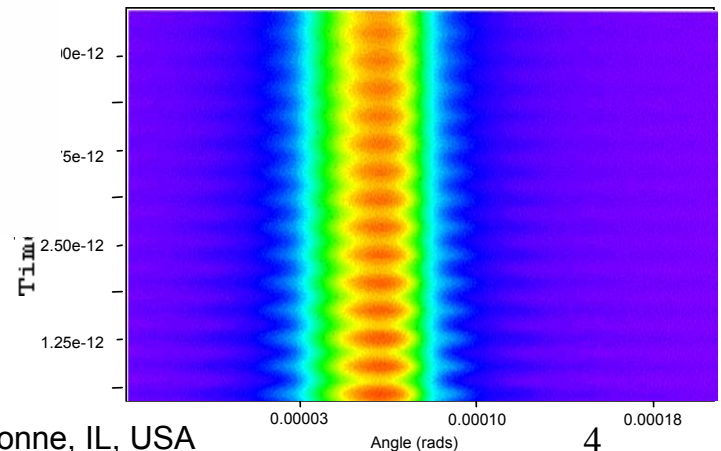

Optical Reflectivity

DeCamp *et al. PRB*, **64** 092301 (2001).
Wark *et al.* SPIE, volume 4143, 2001.

X-ray simulation

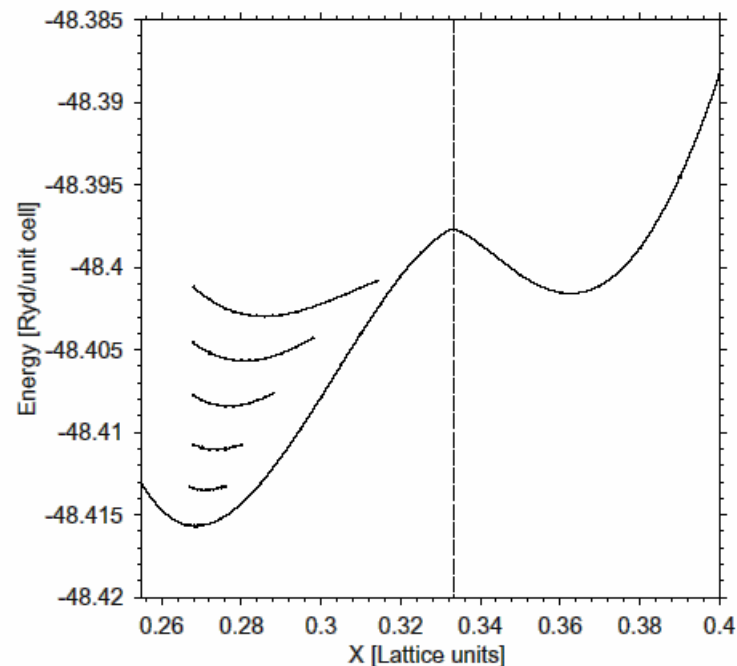SPring8 Workshop, ANL, Argonne, IL, USA
June 2-3, 2003

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D.A. Reis

Simulation of Tellurium Reflectivity Oscillations

(1) First-principles electronic structure theory:

Crystal Energy vs. A_1 Phonon Coordinate

[P. Tangney and S. Fahy, PRL **82**, 4340 (1999);
PRB **65**, 054302 (2002)]

E. Dufresne

June 2-3, 2003

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Comparison of Ultrafast X-ray sources

Laser plasma

- "Perfect" synchronization
- <500 fs
- Low flux
- Low brightness (4π)
- Limited tuning range
- Limited pump-probe delay

ID (3rd Generation)

- picosecond synchronization
- 100 ps typical
- High flux
- High brightness
- Tuneable
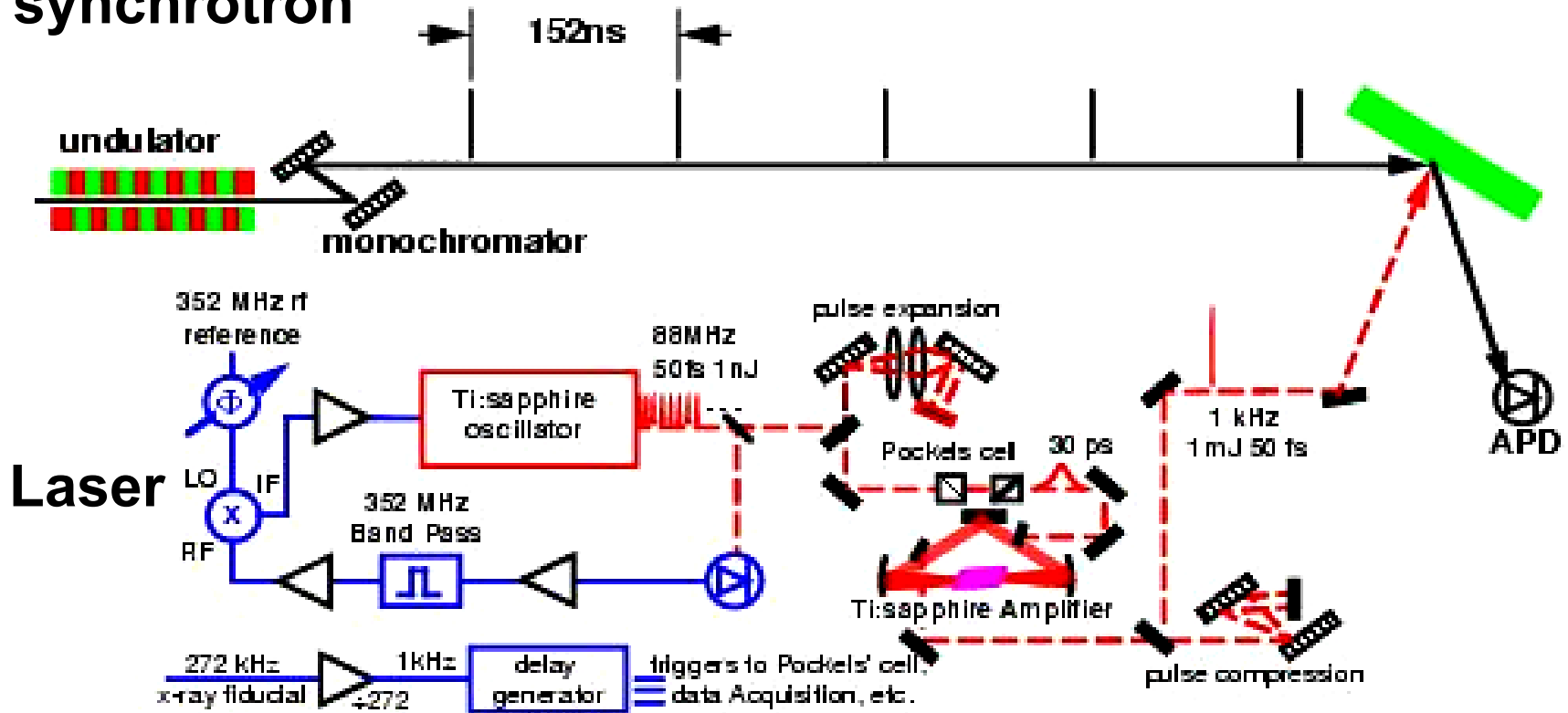- Arbitrary pump-probe delay

Some Ultrafast X-ray Physics activities at the APS

- EXAFS of semiconductors near the laser-melting threshold. (20ID)

- Ultrafast X-ray Spectroscopy of semiconductors (B. Adams et al. APS)

- Streak camera development (Wang et al APS, Reis et al).

- Ponderomotive near edge effects in Kr gas (Young et al. ANL Physics)

- Coherent control of X-ray pulse width: Bragg switch (D.Reis et al 7ID).

Laser-pump–X-ray-probe at 7ID

- Resolution limited by the bunch duration (or the timing jitter)
- Arbitrary pump-probe delay from 18ps-ms

E. DufresneAPS,ESRF,SPring8 Workshop, ANL, Argonne, IL, USA
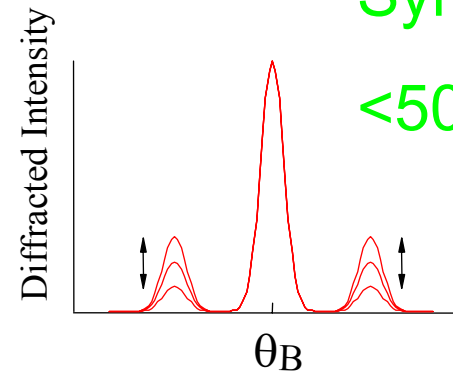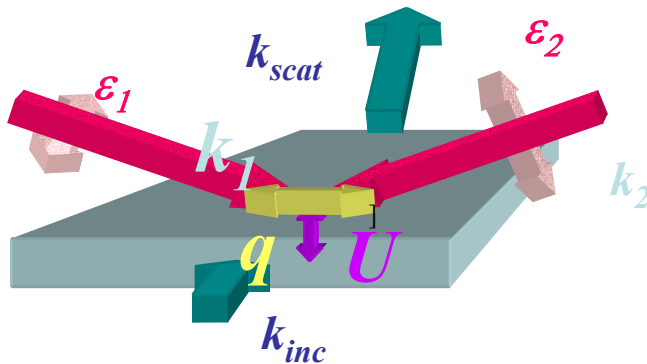June 2-3, 2003

Ultrafast control of x-rays: transient optics

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Transient Grating: Create coherent superlattice using optical phonons
and change the Bragg condition

ε_1 ε_2 k_{scat} k_1 k_2 q U k_{inc}

Buicksbaum and Merlin, S.S. Comm. **111,** 535 1999

"Perfect" Synchronization

<50 fs possible

Diffracted Intensity

θ_B

Turn on reflection with symmetry breaking "zero-wavector" coherent optical phonons

Lattice oscillation up to 10^{-2} have been measured using optical techniques
DeCamp et al. *PRB*, **64** 092301 (2001).
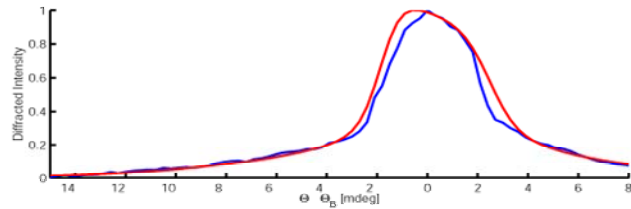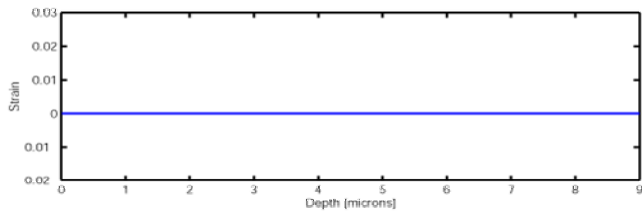Hase et al. *PRL*, **88** 067401 (2002).

Early results in x-ray diffraction
Sokolowski-Tinton et al., CLEO (2002)
(Nature 422, 287 (2003)).

Time (s)

5.00e-12
3.75e-12
2.50e-12
1.25e-12

0.00003   0.00010   0.00018
Angle (rads)

0.1   0.2   0.4   0.5
Reflected Intensity

Time-resolved Bragg Diffraction
(laser pump/x-ray probe)

Impulsive Strain Generation
(Thomsen *et al*. Phys Rev. B (24) 1986.)

- Response of condensed matter to coherent excitation
- Thermal and non-thermal strain generation and melting in semicondu
- Time-domain phonon spectroscopy (inelastic x-ray scattering <meV
- Acoustic propagation and impedances at boundaries
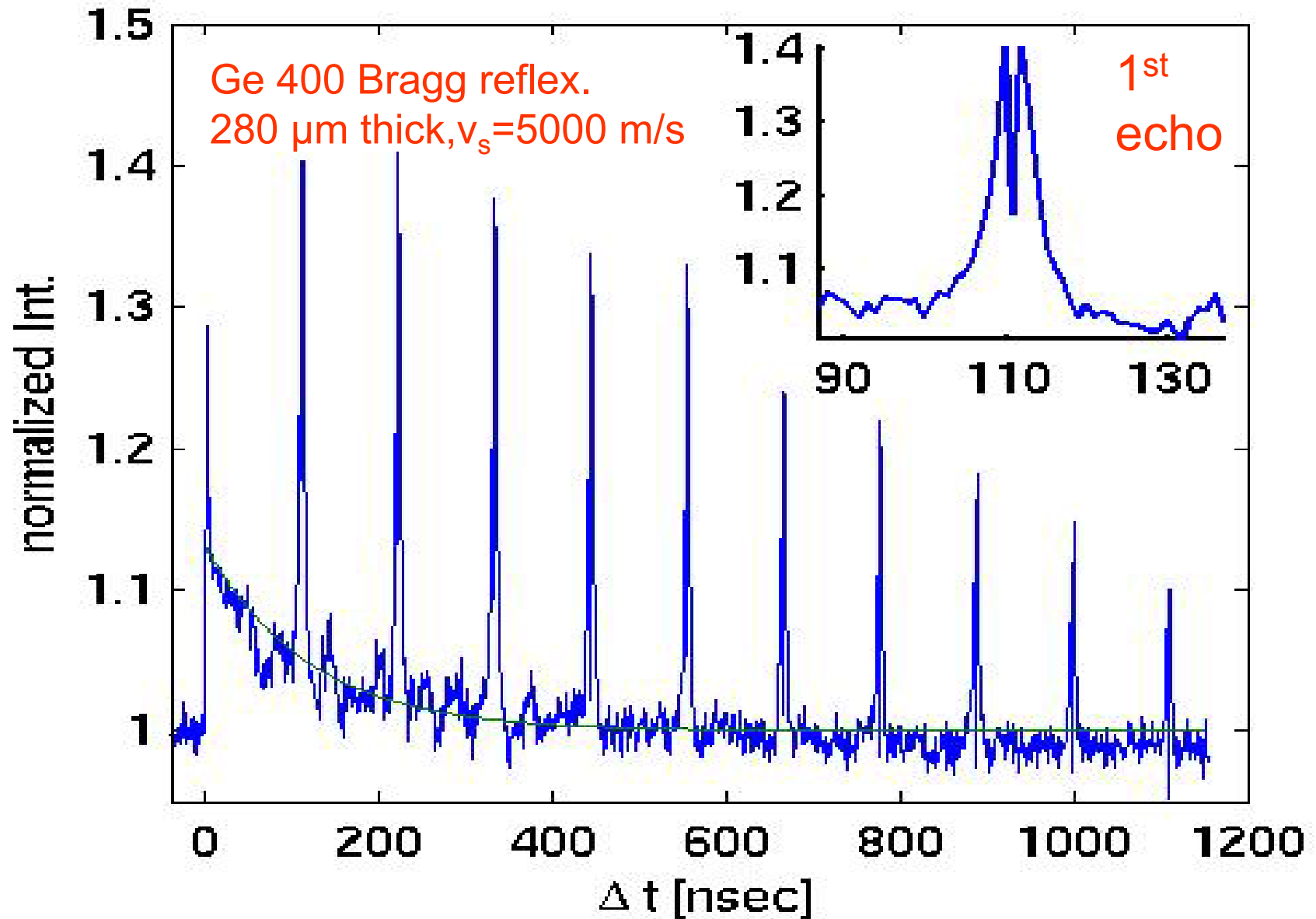- Structural Phase Transitions
- Chemical reactions on surfaces

experiment: InSb 111, 10mJ/cm^2
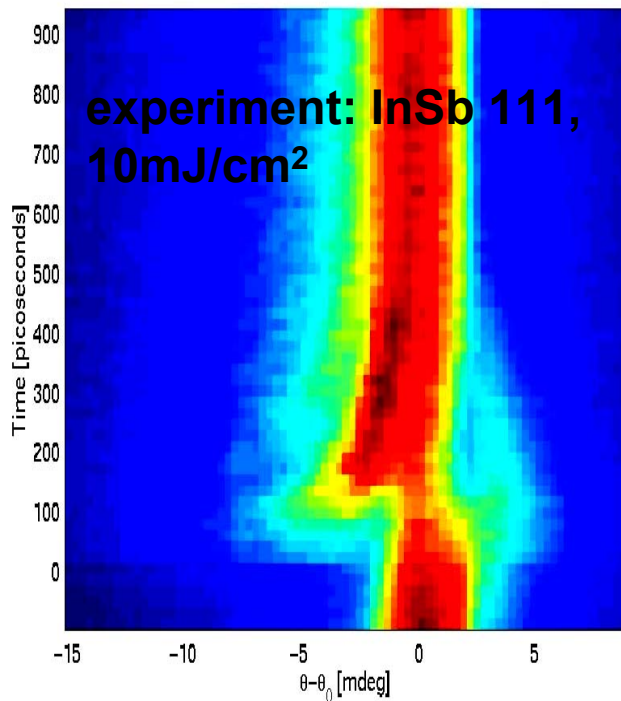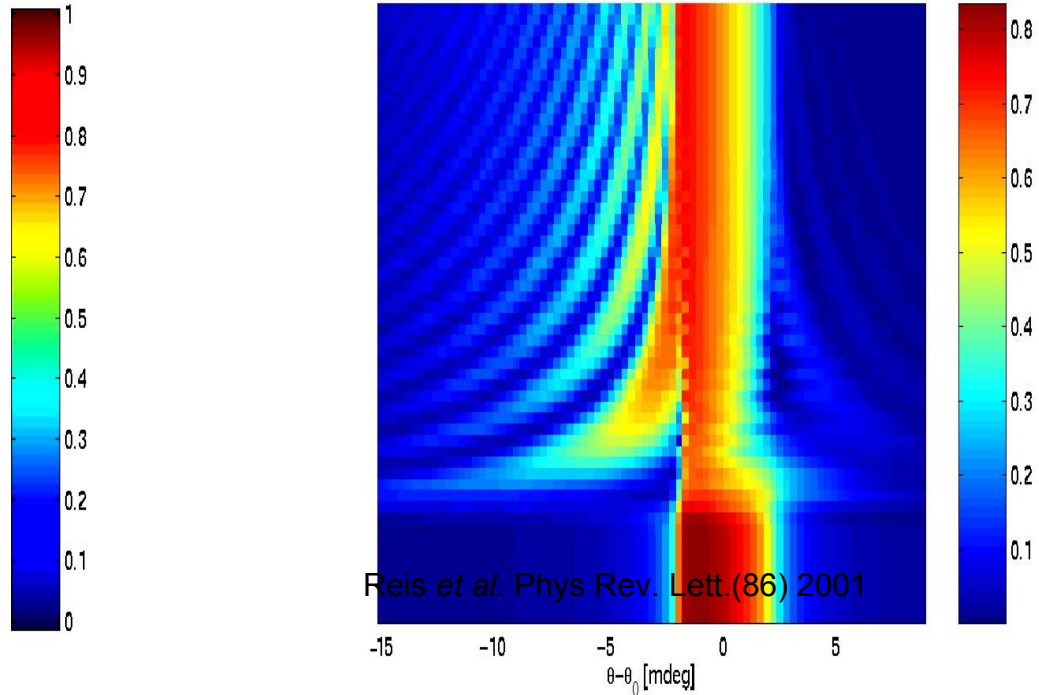
Reis *et al*. Phys Rev. Lett.(86) 2001

acoustic echoes and thermal diffusion

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Ge 400 Bragg reflex.
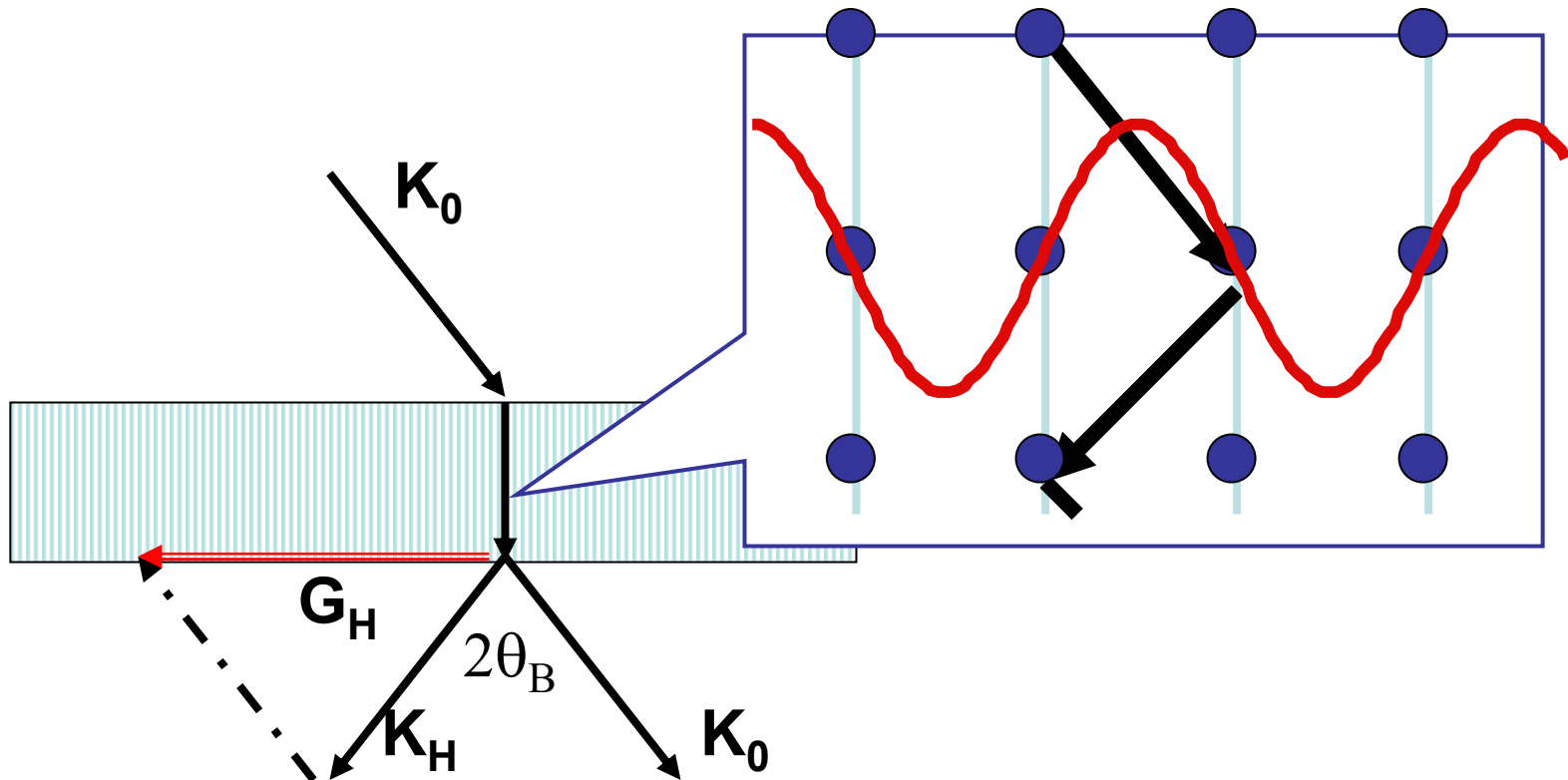280 µm thick, v_s=5000 m/s

1st echo

Borrmann Effect:

(2 r's, 2 n's, 2 f's, 2 e's)
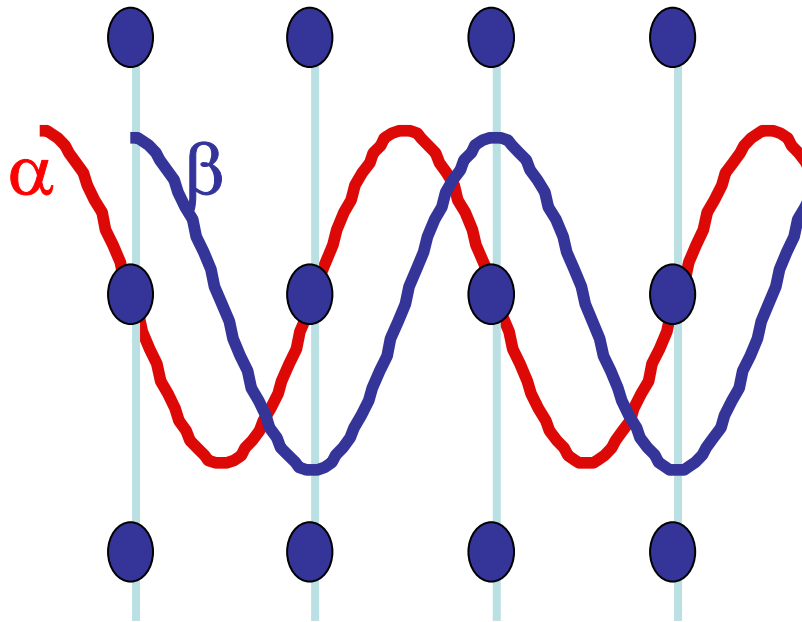an x-ray waveguide in the Laue geometry

"Deflected-Diffracted"   "Forward-Diffracted"
anomalous transmission, or channeling

Pendellösung Effect
(Pendulum like solution)

In Laue geometry: 2 eigenmodes
α – anomalous transmission
β – enhanced absorption

forward and deflected beams:
- linear combinations of α & β:
- mutually coherent

α & β propagate with different v_ϕ

I_0, I_H beat with wavelength $\Lambda = (K_\alpha - K_\beta)^{-1}$

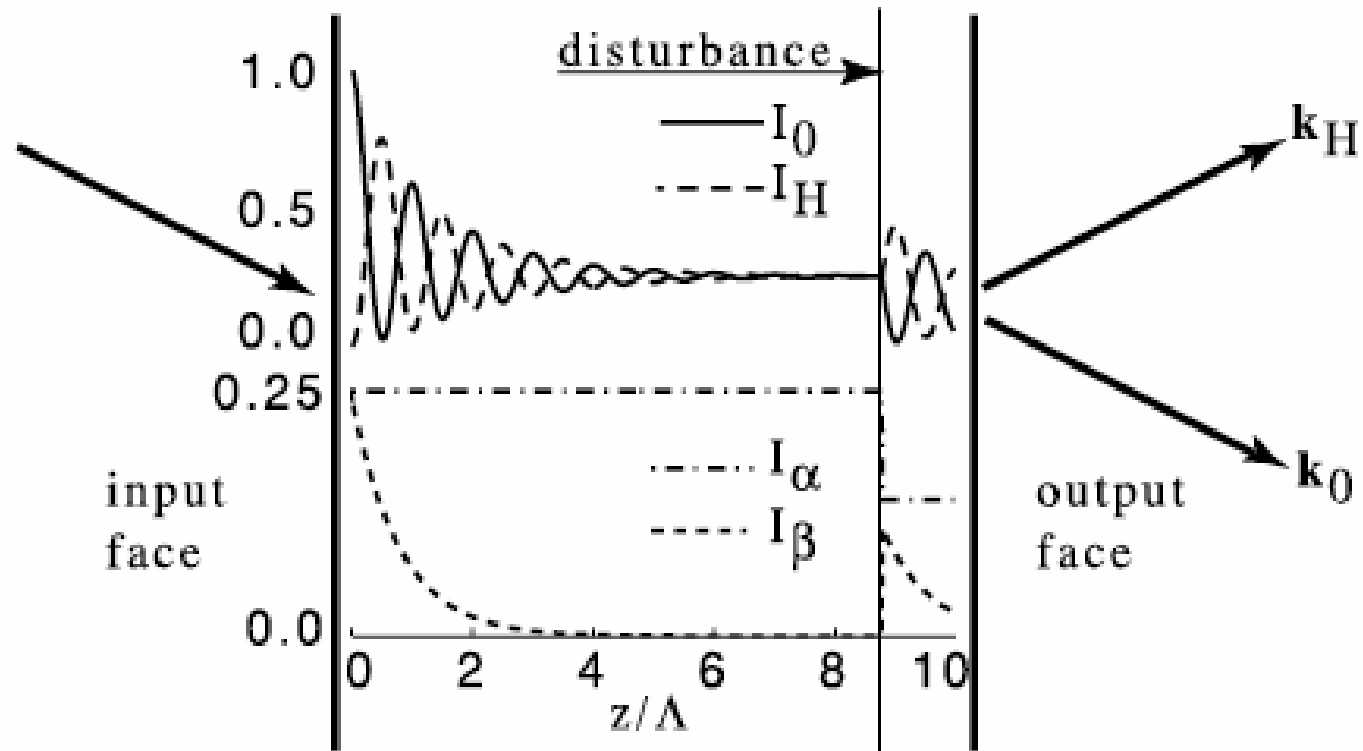$$I_0 = \left| E_\alpha \, e^{i\vec{K}_\alpha \cdot \vec{r}} + E_\beta \, e^{i\vec{K}_\beta \cdot \vec{r}} \right|^2$$

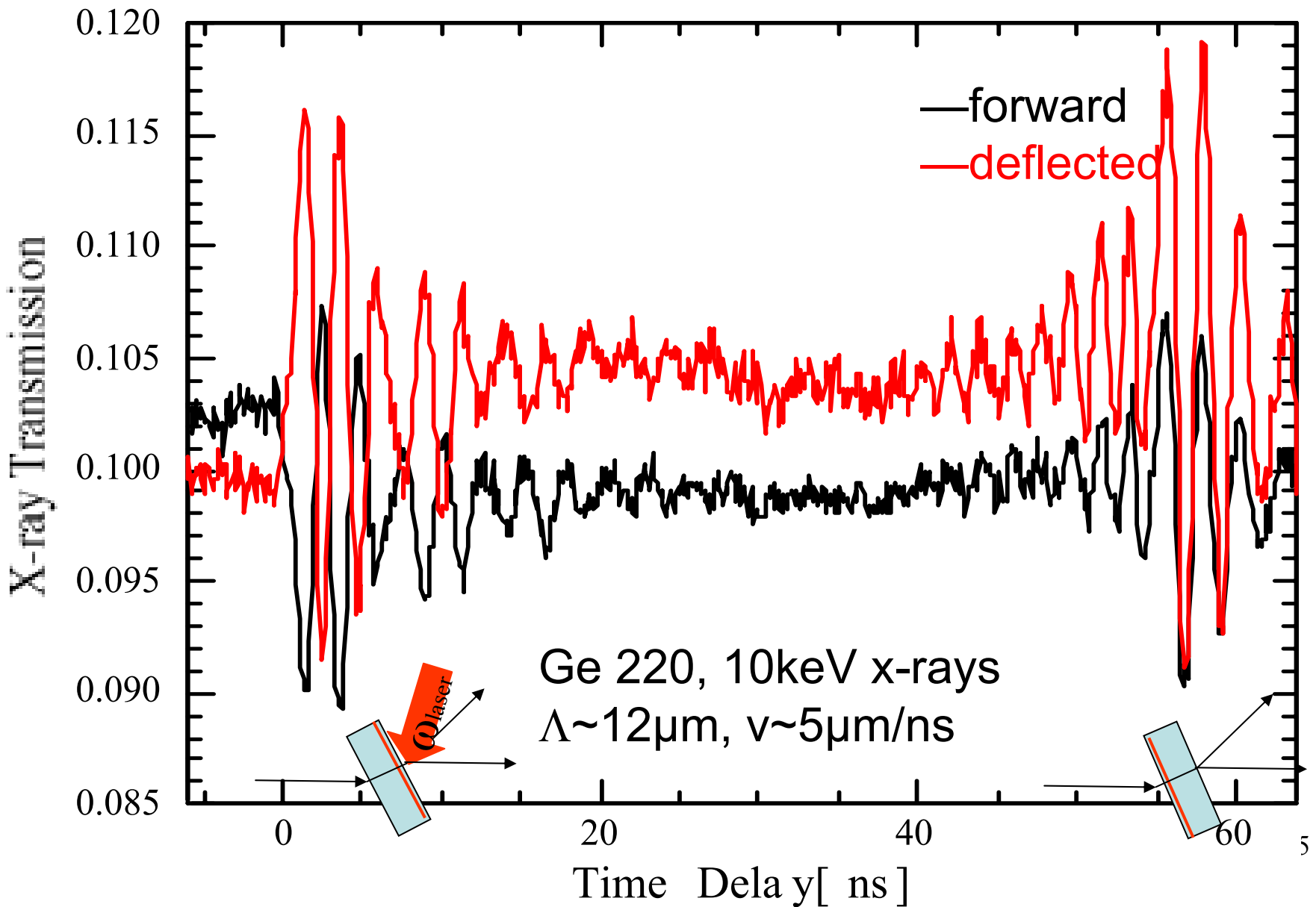$$I_H = \left| E_\alpha \, e^{i\vec{K}_\alpha \cdot \vec{r}} - E_\beta \, e^{i\vec{K}_\beta \cdot \vec{r}} \right|^2$$

Acoustic Pulse, acts as moving lattice disturbance, regenerates β

Programmable X-ray Beamsplitter

Coherent control of pulsed X-ray beams

M. F. DeCamp*, D. A. Reis*, P. H. Bucksbaum*, B. Adams†,
J. M. Caraher*, R. Clarke*, C. W. S. Conover‡, E. M. Dufresne*, R. Merlin*,
V. Stoica* & J. K. Wahlstrand*

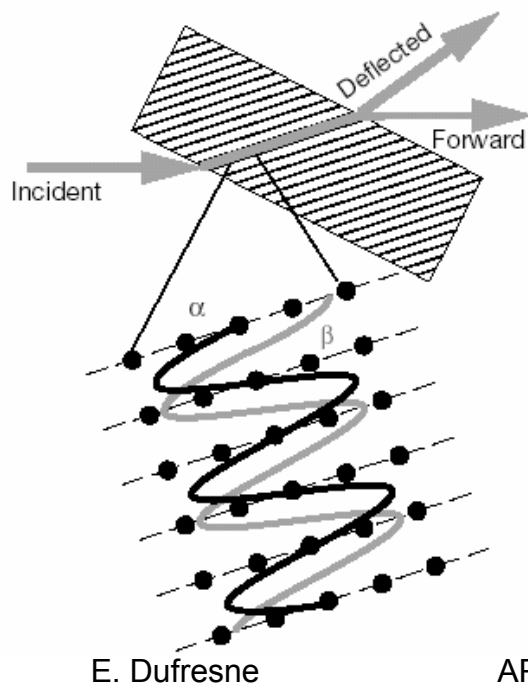* Department of Physics and FOCUS Center, University of Michigan, Ann Arbor, Michigan 48109, USA
† Advanced Photon Source, Argonne National Labs, Argonne, Illinois 60439, USA
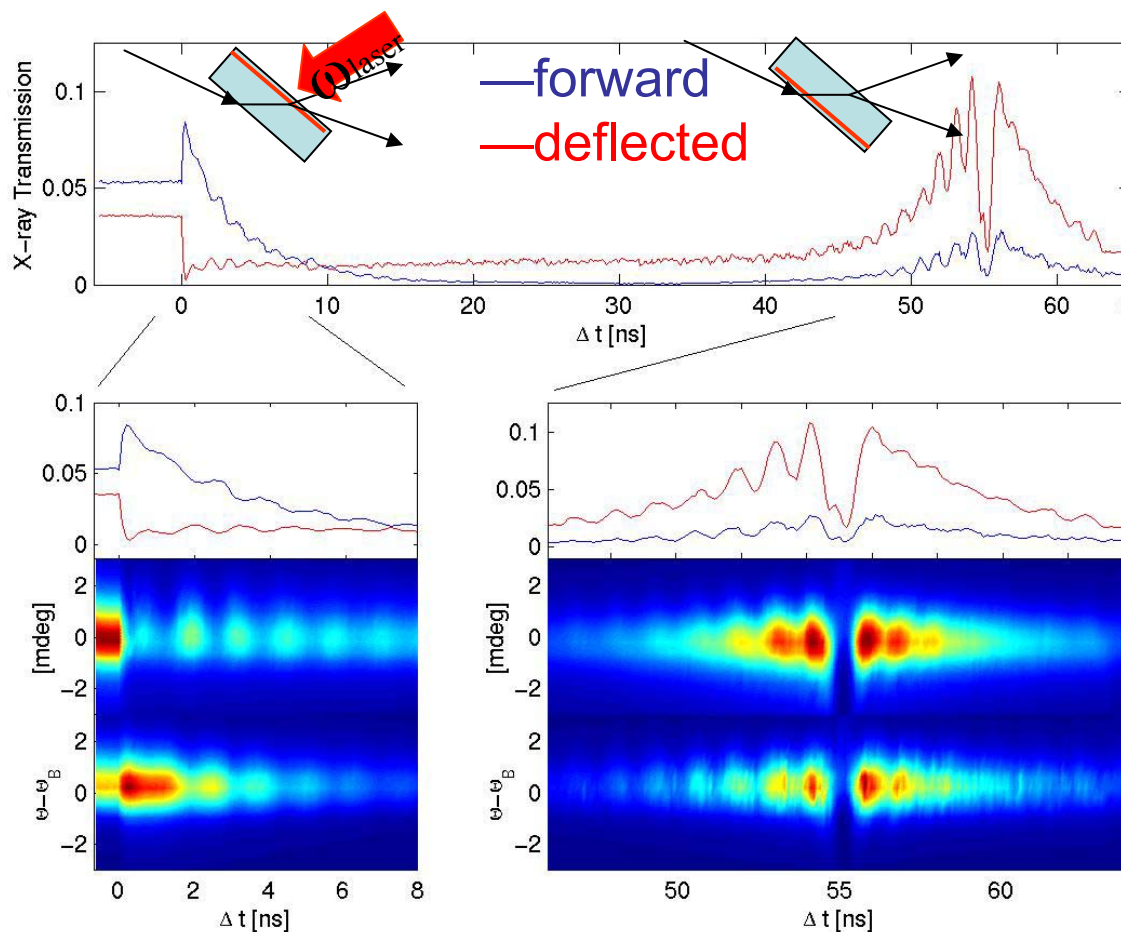‡ Colby College, Waterville, Maine 04901, USA

•Uses coherent acoustic pulse to control the intensity and direction of transmitted x-rays

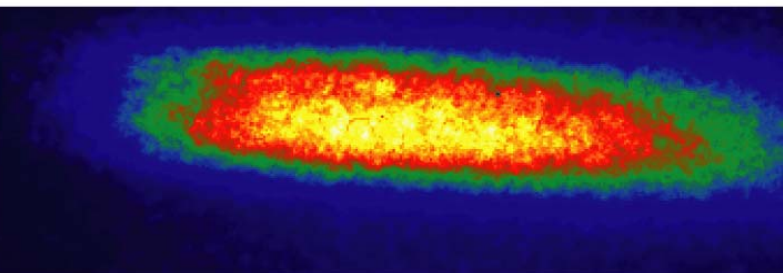•fast transient not limited by acoustic propagation

E. Dufresne   APS,ES

streak camera resolves transient switch
~6 times the speed of sound.

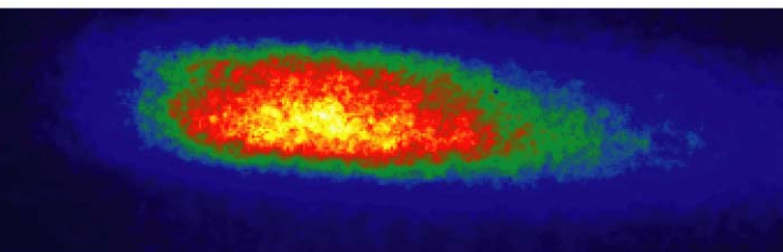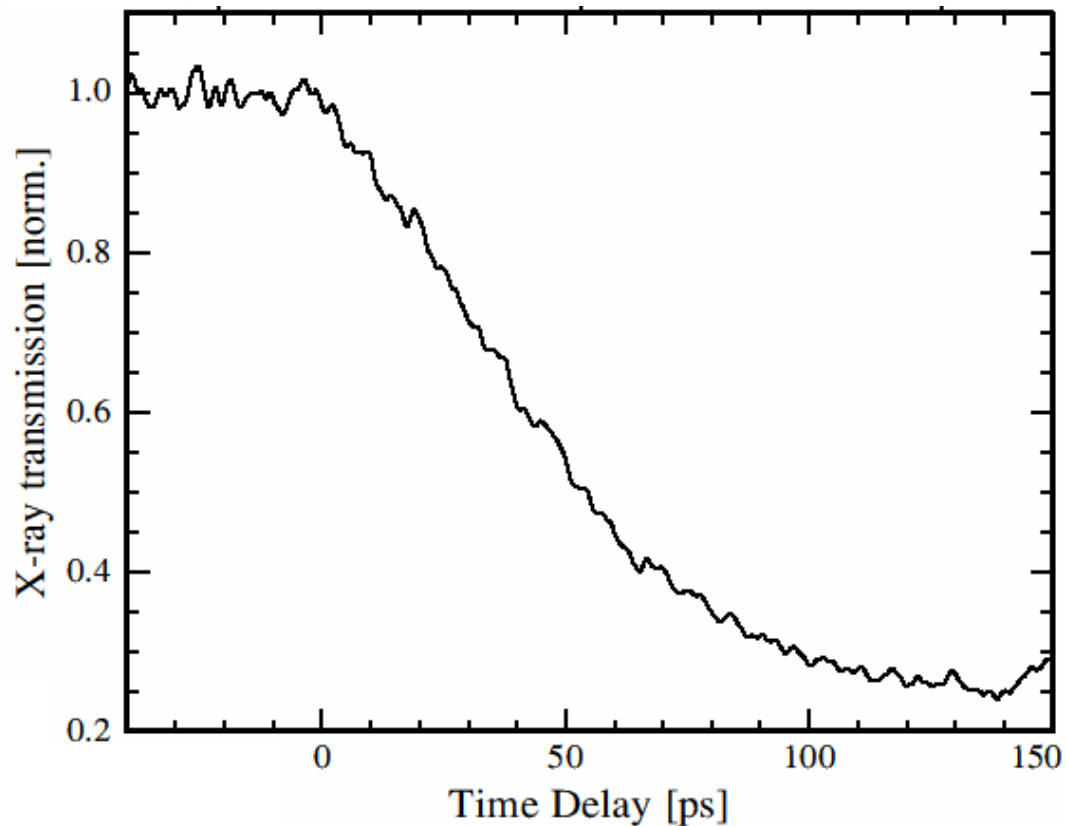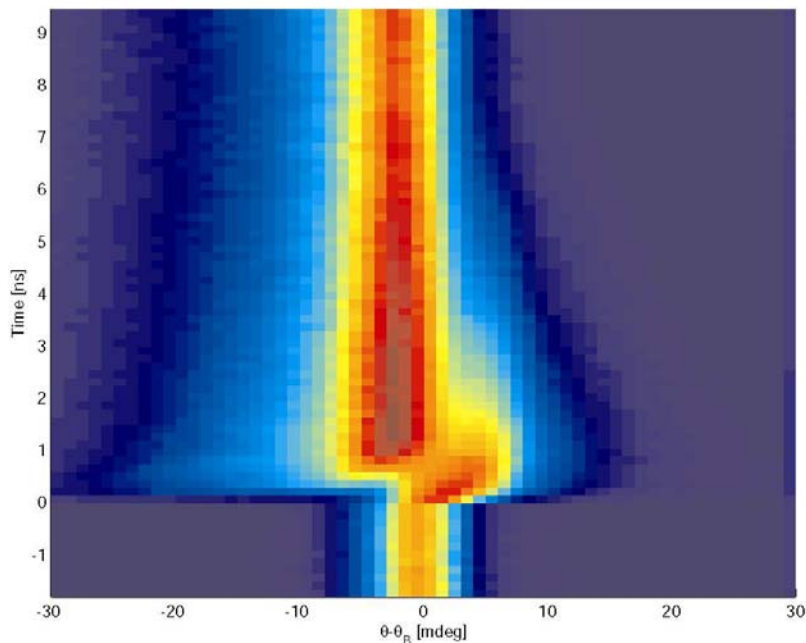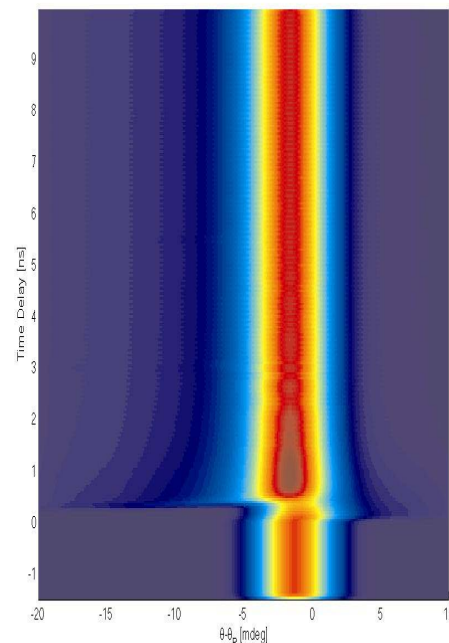Laser off

Laser on

From Bragg diffraction we know that thermoelastic model not sufficient
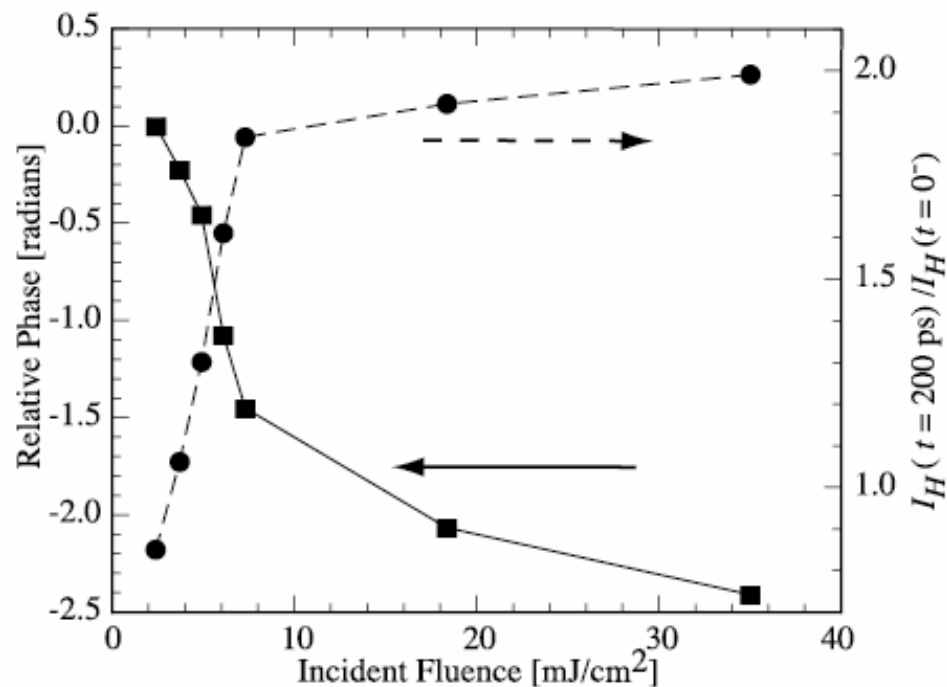
Ge 400 symmetric Bragg
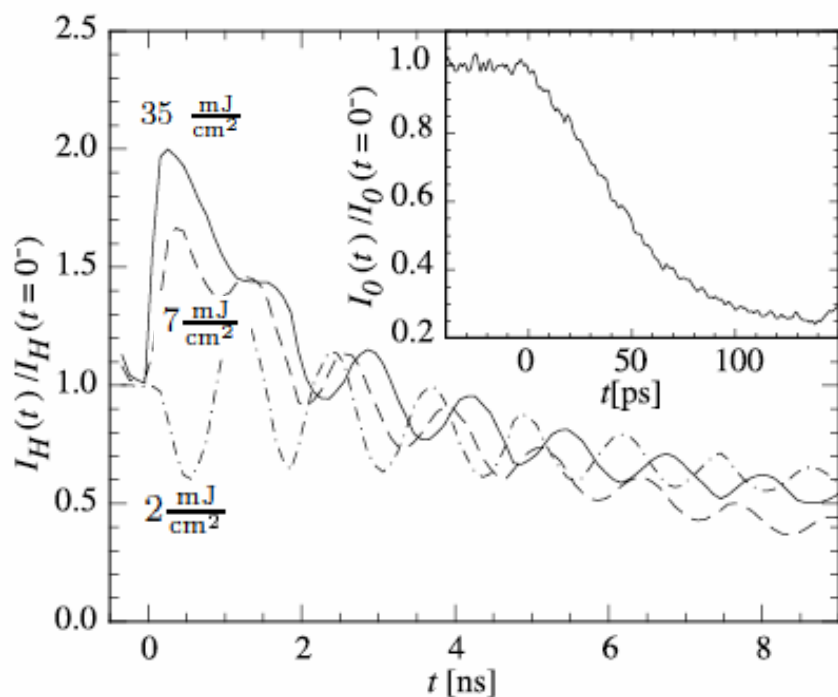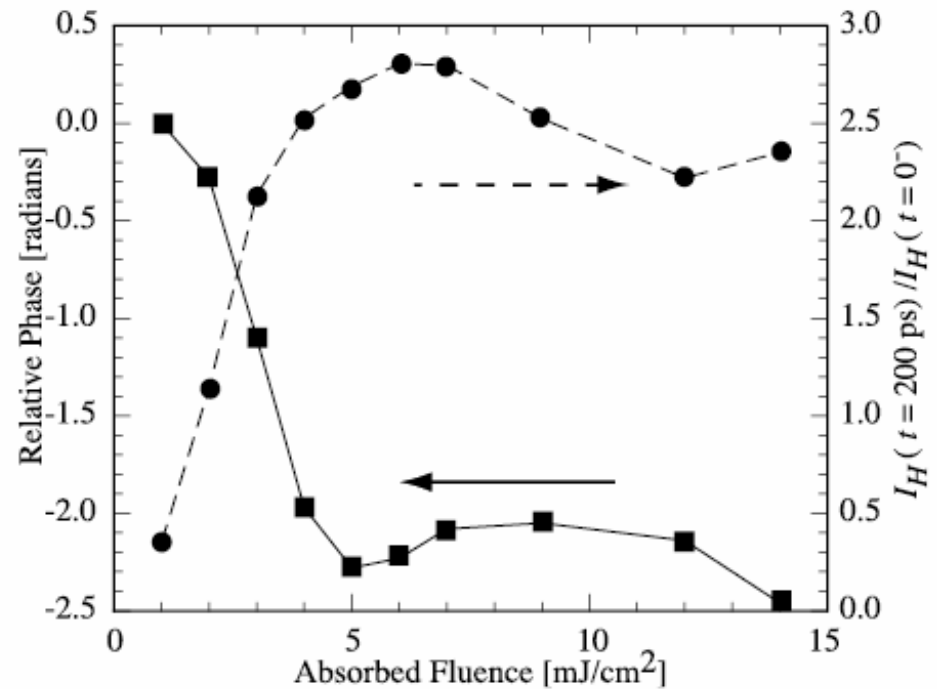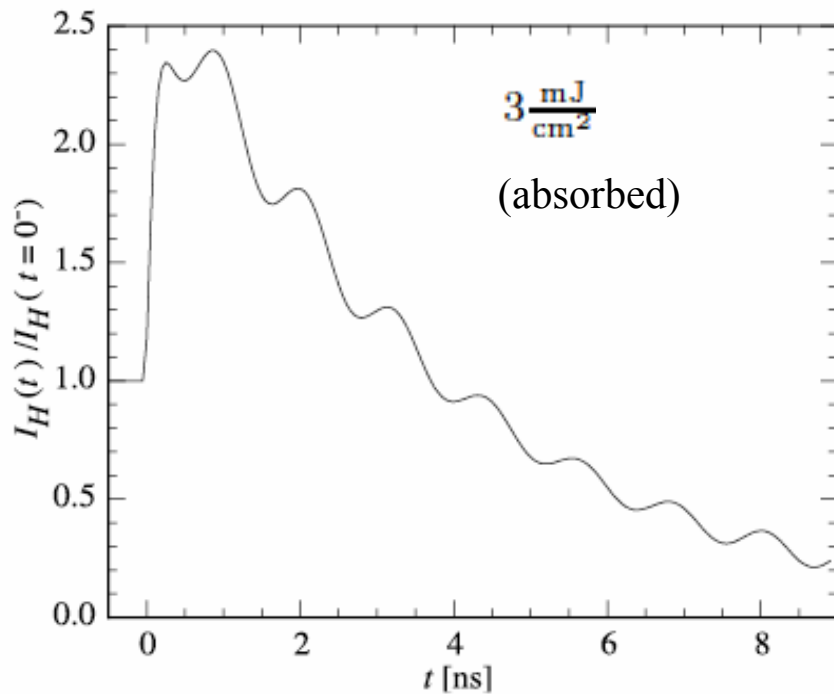

Data

simulation

Fluence dependence of transient, phase

cond-mat/0301002

Simulations results

Probe of bulk carrier dynamics

cond-mat/0301002

Conclusions

Impulsively generated coherent acoustic phonons are novel non-equilibrium states of matter which can modulate X-ray diffraction.

-In Bragg geometry: Detailed ps strain determination can be measured as well as the phonon dispersion and attenuation.

-In Laue geometry: One can track the phonon propagation through the entire bulk. This allows for ns coherent control of x-ray pulses. Well separated beams make a convenient and efficient switch. Detailed plasma generated strain models are necessary to explain the fast transient.

Scaling the technique to coherent optical phonon excitations should lead to fs control.

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